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STUDIES ON SOME INDIAN VEGETABLE OILS. PART V. TEMPERATURE EFFECT ON GAS ABSORPTION AND OTHER PHYSICAL PROPERTIES

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ABSTRACT. The absorption of air, hydrogen, nitrogen and carbon dioxide by castor oil at different temperatures within the range 20°C—90°C has been measured by the manometric method and the Bunsen absorption coefficients determined. It has been observed that for all the gases there is a certain temperature at which the Bunsen coefficient attains a maximum value, and a mechanism of association between the gas and liquid molecules is suggested to explain the phenomena.

The values of viscosity at different temperatures within the same range have been measured in absolute units by the capillary method suggested by Gemant. The values obtained have been found not to satisfy the logarithmic relation of Andrade, and the logarithms of viscosity have been fitted into an equation of the form $\log\eta = \log A - \alpha t + \beta t^2 - \gamma t^3 + \delta t^4$ and the difference between observed and calculated values determined.

The variation of refractive index with temperature within the range 0°C—70°C has been determined and the variation is found to be a linear one.

INTRODUCTION

The present investigation deals with the variation with temperature of the coefficients of absorption of air, H_2, N_2 and CO_2 by castor oil and of other physical properties, such as viscosity, density, refractive index, dielectric constant, etc., within the working range of temperature, *viz.*, 20°C—90°C.

In the International Critical Tables [1] one finds considerable amount of literature on the absorption of gases by inorganic and organic liquids. The only data on the absorption of gases by mineral oils are those of A. Gemant [2] and F. M. Clark,[3] but very little work has been done on the gas absorption and other physical properties of vegetable oils at different temperatures. The only work on the same line on vegetable oils is that of G. N. Bhattacharyya,[4] who measured the absorption of air by some Indian vegetable oils at room temperatures only. Gemant in his measurement of gas absorption by mineral oils found that the absorbed amount of air per unit volume of mineral oil remained practically constant at 20°C and 80°C. He, however, gives no account of the absorption at intermediate temperatures. Clark, on the other hand, has shown that there is a definite increase in the absorption coefficient (of about 10%), within the above range of temperature ; but in his graph representing the variation of

absorption with temperature, one notices that there is a tendency towards a decrease in the absorption for higher temperatures.

In connection with drying and insulating oils the investigations on their coefficient of gas absorption have an important bearing. F. M. Clark [5] has shown that the dielectric strength of oils is dependent on the nature and amounts of dissolved gases. He also suggests that most theories of breakdown of solid dielectric impregnated with oil are not satisfactory because of the neglect of this gas-absorption factor. Quite recently Race,[6] while discussing the tests on oil-impregnated paper, draws attention to the different causes of electric breakdown of impregnated paper insulation as preceded by gaseous ionisation. He states that in a nominally gas-free liquid-filled system the dielectric strength should increase with the degree of degasification. Copelmann and Gyemant [7] supposes an electrode layer of high stress due to space charge acting on a layer of adsorbed gas, thus creating gas pockets or filaments leading to gaseous ionisation and breakdown and one would expect that in the case of liquids with a tendency of high absorbing power for gases the dielectric strength will suffer.

Hill[8] has considered the phenomena of heterogeneous equilibrium between gas and liquid according to the Distribution Law and the Phase Rule, and has discussed Henry's Law and its modifications due to association and dissociation. But the effect of temperature on the absorption of gases has not been discussed.

The use of vegetable oils as solvents of paints and as lubricants are generally known, but sufficient data as regards their other physical properties are not available, though a fair amount of data is available in the case of mineral oils.

Hence, in order to investigate the applicability of vegetable oils for the purposes in which mineral oils are used, it is of interest to have a thorough study of all the above-mentioned properties. In almost all these applications the oils have to work within a certain range of temperature, and hence a study of the effect of variation of temperature within this range on these properties is of importance. It is, also, worthwhile to have an idea of the mechanism of absorption of gases and the variation of the coefficients of absorption in oils within the same range of temperature.

An account of the investigations so far carried out on vegetable oils has already been given by G. N. Bhattacharyya[9] of this laboratory. The same author published the results of the variation of viscosity of several vegetable oils with temperature and his results are in Redwood units and Fahrenheit degrees. It is thought worthwhile to measure the viscosity in absolute units to enable one to have an idea of the mechanism of viscous forces acting in the case of vegetable oils.

THE MECHANISM OF ABSORPTION OF GASES BY LIQUIDS

In order to form a mental picture of the mechanism of gas absorption in liquids, one can imagine the molecules of gas, enclosed in a chamber containing

a liquid to enter into the liquid surface during their random motions by virtue of their kinetic energy. Having entered the liquid, these molecules move from one position to another, and in their motion any of the following incidents may take place : they may come in close proximity of a liquid molecule, as a result of which the two may form a combination, executing motions as a whole ; or they may come into collision with each other or with combinations of the first kind and thereby knock off the gas molecules with sufficient energy to drive them out of the liquid altogether ; or, thirdly, they may in their motion come out of the liquid without encountering any other molecule.

One can, therefore, assume that on an average a number of gas molecules are in association, so to say, with the liquid molecules, which are supposed to be in a state of oscillation about a slowly displaced position of equilibrium ; the total number of such molecules depending on the pressure of the gas, the nature of the liquid and gas molecules and the prevailing temperature. So that at any instant there are a number of liquid and gas molecules associating together to form a juxtaposed type of molecule.

One can imagine that a number n_1 of the free gas molecules is entering into the liquid at any instant, while a number n_2 is coming out of the liquid. At a certain stage, for a particular temperature, n_1 becomes equal to n_2 and an equilibrium is reached, and the number of molecules confined within the liquid in the above associated state measures the coefficient of absorption.

The association or mutual cohesion between the two molecules can take place due to anyone of the several types of forces, such as, van der Waal attractive forces (otherwise known as dynamic polarisation), electrostatic forces due to permanent dipoles, ionic forces, polarisation forces. Of these, the energy due to van der Waal forces have been found to vary inversely as the sixth power of the distance between the molecules, whereas the forces between dipoles vary inversely as the cube of the distance. It may be either or both of the first two types, but since the second type, *viz.*, the force between dipoles is stronger and the type of liquids under consideration are known to have a chain-like structure, it is reasonable to suppose that the latter type of force preponderates in the phenomenon of gas absorption.

One can thus picture such an associated molecule having a certain mutual potential energy E, executing oscillations about a slowly displaced position of equilibrium. The thermal agitation due to variation of temperature as also the change in intermolecular forces in the gas due to variation of pressure will, therefore, interfere with this phenomenon of association. The magnitude of the absorption is thus dependent on the value of this mutual potential energy E, while the variation of the coefficient of absorption with temperature will be governed by the fraction of the total number of molecules possessing this energy E. At a definite pressure the probability of association of the gas and the liquid molecules will be maximum at a certain temperature. For lower or higher tem-

peratures there would be fewer molecules in the associated state. There is thus an equilibrium condition at a definite temperature between the molecules of the gas and the liquid-gas associated groups similar to that which occurs between different phases of a substance which can pass from one phase to the other.

In deriving the expression for Wien's law for black-body radiation Lorentz[10] has pointed out that the equilibrium between rays of different frequencies as affected by temperature is analogous to the equilibrium condition which obtains between different phases of a substance which can pass from one phase to the other.

In the case of Wien's law the energy of radiation is a function of frequency and temperature and an exponential relationship between these factors are operative. Extending the analogy to the present case, one would expect that the energy of association is governed by an exponential relation between temperature and pressure and the corresponding oscillatory energies of gas and liquid molecules.

Thus for a pressure P and temperature T the energy

$$E = C_1 T^{-a} e^{-\frac{C_2}{TP}}.$$

The values of the constants C_1, C_2, a, would depend on the oscillation frequencies of gas and liquid molecules, the nature of permanent dipole moments and induced moments of the molecules and the Boltzmann constant. These factors are difficult to reckon due to the uncertainty of our knowledge of the actual magnitudes of the forces and the actual expression for the interactive forces in operation.

VISCOSITY AND ITS VARIATION WITH TEMPERATURE

The mechanism of the viscosity of liquids has been the object of investigation since Newton who conceived the existence of a shearing stress at any point of a liquid moving in parallel layers. Since Newton, the same question has been discussed by various investigators from different physical aspects, which has been described by G. N. Bhattacharya[9] in his paper on the Viscosity of Vegetable Oils.

Andrade[11] has treated the theory of viscosity from the point of view of the communication of momentum from layer to layer taking place at the extreme libration of molecules in each layer oscillating about a very slowly displaced position of equilibrium. He considered the variation of viscosity with temperature as due to the effect of change of temperature on this interchange of momentum. He considered the change of potential energy of the system, due to change in temperature, in the light of Boltzmann's distribution law, and arrived at the relation

$$\eta_1 = Ae^{C/T}$$

where A and C are constants.

He next considered the effect of temperature on the volume of the liquid, and on the distance between the molecules, since the distance between the molecules increases as $v^{1/3}$ and the number of molecules per unit area diminishes as $v^{2/3}$ where v is the specific volume of the liquid, the decrease in viscosity due to this effect alone is as $v^{-1/3}$. He further assumes that the potential energy involved in the condition of communication will be a function of the volume, $f(v)$, so that, taking all these effects into consideration, the relation changes to

$$\eta_1 = Ae^{cf(v)/T} \cdot v^{-1/3} .$$

Since the average potential energy of a molecule is to a first approximation given by a/v in van der Waal's equation, it is supposed that the part of the potential energy considered in the case of viscosity varies in a similar way, and hence the relation can be taken as

$$\eta_T = Ae^{c/vT} \cdot v^{-\frac{1}{3}}$$

or
$$\eta_T . v^{\frac{1}{3}} = Ae^{c/vT}$$

or
$$\log_e \eta_T = \log_e A + \frac{c}{vT} - \tfrac{1}{3} \log_e v.$$

Whereas Andrade starts on the basis of a static equilibrium between the forces exerted by the various molecules on each other, van der Waals (Jr.)[12] has treated the theory of viscosity in the same light of transportation of momentum due to collision, but has based his theory on the consideration of heat motion from the start.

He obtained the relation

$$\eta = \frac{2\sqrt{2\pi}}{15} n^2 d^4 ma. \; e^{-E/RT} \; e^{-b/v}$$

where d is the diameter of a molecule ; $a\sqrt{3/2}$ is the quadratic mean velocity of the molecule ; b is the quantity occurring in the equation of state which is connected with the total volume of the molecules contained in volume v ; and E is the difference between the mean potential energy of the molecules in the liquid, and the potential energy at the moment of collision (calculated per mole).

This relation can be transformed into the form in which Andrade expressed the relation for viscosity by considering every molecule in a liquid as being surrounded by a set of " first neighbours," and thus is practically enclosed in a sort of cavity of irregular form, not much greater than the molecule itself.

From the above relation it will be seen that the change of viscosity with temperature is influenced mostly by the factor $(1 - b/v)$, and by a which is $\sim \sqrt{T}$, n the number of molecules per unit volume and the exponential factor.

Frenkel [13] worked out an expression on the basis of approximate equality of the specific heat of the same substance in the liquid state at low temperature and in the solid state. Batschinski,[14] Macleod,[15] and Lederer[16] found similar expressions all on the assumption that η is primarily a function of the specific volume.

All the different theories have been fully discussed by the Committee for the study of viscosity of the Academy of Sciences at Amsterdam,[17] and it would seem that the relation between viscosity and specific volume of a liquid is of primary importance and that the particular dependence of the viscosity upon temperature should for a large part be explained as a secondary effect, to be derived from the relation between specific volume and temperature. Though the experimental results are in agreement in a number of cases with the different expressions stated above it is found that in considerable number of liquids, specially the oils, the theoretical expressions are not in conformity with the experimental results.

In this connection it might be of interest to note that in the final expression deduced by Andrade, *viz.*, $\eta v^{\frac{1}{3}} = Ae^{\frac{c}{vT}}$ the effect of temperature on the potential energy has not been taken into account. An attempt has been made therefore to consider the effect of temperature on the van der Waal's constant a/v. One can express the relation as

$$\log \eta = \log A - \alpha t + \beta t^2 - \gamma t^3 + \delta t^4 - \ldots\ldots\ldots$$

This has been done by fitting the logarithmic curve by the method of least squares.

EXPERIMENTAL

For the measurement of co-efficient of absorption of gases, the same manometric method as described by G. N. Bhattacharyya[4] was utilised. To obtain measurements at different temperatures, the flask containing the oil was kept completely enclosed in a thermostatic arrangement. A box, the inner wall of which was provided with resistance wires for heating purposes, was suitably lagged for maintenance of uniform temperature inside it, and by means of variable resistances used in series with the heating coils the temperature inside the box could be regulated very nicely. Hydrogen was obtained by electrolysis from water, and was dried by passing through a series of calcium chloride towers before being passed into the flask. Nitrogen gas was obtained by the action of

a solution of ammonia and ammonium carbonate on copper turnings by the method described by C. V. Brunt.[18] The gas, before collection, was passed through wash towers containing solutions of sulphuric acid, pyrogallic acid and caustic potash for the removal of impurities, such as ammonia, oxygen and carbon dioxide. Before being passed into the flask it was again passed through a second set of wash bottles containing the same solutions and then through calcium chloride towers. Carbon dioxide gas was purchased in cylinder and was stored in aspirator jars for controlling the pressure of the gas passed into the flask. The gas was passed through towers containing pumice stone soaked with sulphuric acid for the removal of moisture. The Bunsen absorption coefficient was determined in the same way as done by Bhattacharyya. The results obtained are given in Tables II-V.

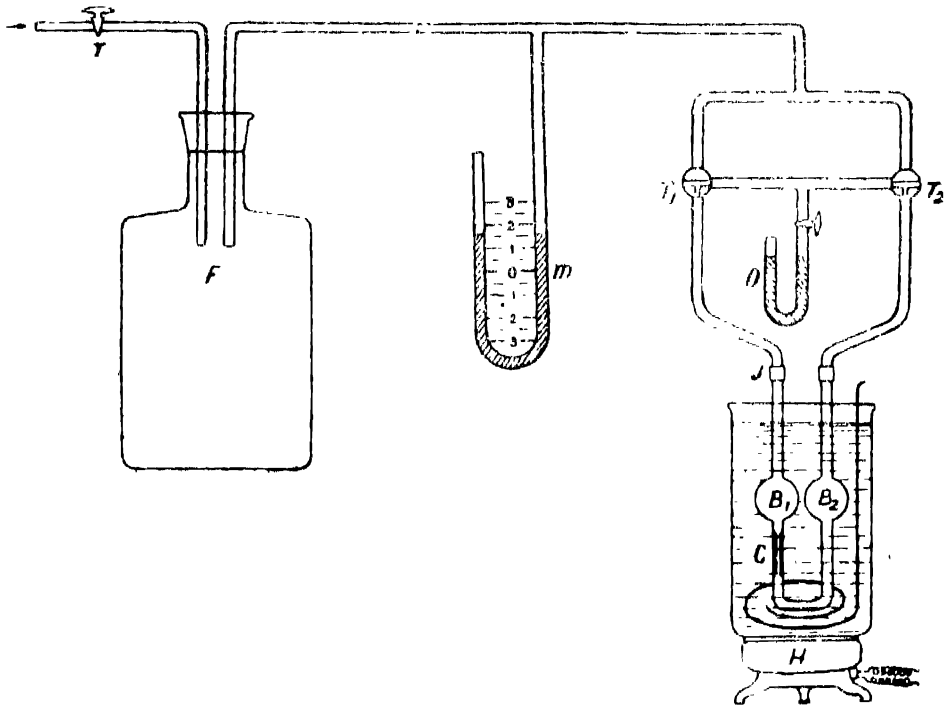

FIGURE 1

The apparatus used for the determination of viscosity in absolute units was as shown in Fig. 1 and was designed in the laboratory after Gemant.[2] As shown in the figure, the apparatus consists of a big flask F which acts as a reservoir of air at a pressure indicated by the mercury manometer *m*, and because of its large capacity the pressure of air remains almost constant during the time taken by the liquid to flow through the capillary tube. The flask is connected to the measuring apparatus through the three-way stop-cocks T_1 and T_2. The measuring apparatus is a U-tube with two bulbs B_1 and B_2 near the top of the

two limbs, the bulb B_2 being slightly bigger than B_1. A portion C in the limb containing B_1 is a capillary tube of length about 10 cms. The two limbs of the U-tube are joined to the three-way stop-cocks T_1 and T_2 by means of rubber tubing at JJ. Two marks at the top and bottom of the bulb B_1 indicates the volume of liquid which flows out of B_1, through the capillary tube C into B_2 Between T_1 and T_2 is connected an oil manometer O,which indicates any small difference in level in the two limbs of the U-tubes. The temperature of measurement could be maintained constant by immersion of the U-tube in a water bath heated electrically.

The length and diameter of the capillary portion C was measured by taking several measurements of the weights of mercury required to fill in different lengths of the capillary and the total weight required to fill up the whole length. The volume of the bulb B_1 between the marks was also determined in a similar way.

The method of procedure consists in thoroughly cleaning and drying the U-tube and then filling it up with a quantity of the oil sufficient to fill up the bulb B_2. Next keeping T_1 in the position where the limb B_1 is open to the atmosphere, the limb B_2 is connected to the reservoir F, in which the air has previously been compressed to a pressure of about 20 cms. of mercury, by means of the stop-cock T_2, and thereby the oil is transferred from the limb B_2 to the limb B_1, the flow being stopped by rotating T_2 into its position for connection of limb B_2 with the atmosphere, when the level of oil in limb B_1 has reached about 2 cm. above the mark. With T_2 in the position mentioned, the level of oil in limb B_1 slowly begins to fall ; when the level has reached the mark above bulb B_1, the stop-cock T_1 is turned so that communication is made between limb B_1 and the reservoir F. Simultaneously with the opening of stop-cock T_1 to the reservoir F, the stop-watch is started and the pressure in the manometer m is noted. Finally, as the liquid column reaches the mark below bulb B_1, the stop-watch is stopped and the pressure in the manometer again noted. The mean of the two readings of the manometer is taken as the effective pressure on the liquid during the period it flows out of B_1. For each temperature several readings are taken with different mean effective pressures to obtain consistent results

According to Poiseulle the relation between the viscosity of a liquid and its flow through capillary tubes at a constant temperature is given as

$$\eta = \frac{P\pi R^4 T}{8LV}.$$

Adding the correction factor when the kinetic energy of the out-flowing liquid is taken into consideration, the relation becomes

$$\eta = \frac{P\pi R^4 T}{8(L+nR)V} - \frac{m\delta V}{8\pi(L+nR)T}$$

where η = the viscosity of the liquid,

P = the effective pressure,
R = radius of capillary,
V = volume of out-flowing liquid in time T,
L = length of the capillary tube of radius R,
n = a certain constant depending on the characteristics of the ends of the tube,
δ = the density of the liquid at the temperature of measurement,
m = a constant.

The value of $n = 1.64$ and $m = 1$. In the case of vegetable oils δ is generally $= 1$. By making L considerably large, and since V/T for oils is naturally small, the second term can be neglected. For the apparatuas in use, since the values V, R and L are constants, by using a liquid of known viscosity, the apparatus can be calibrated such that for any liquid the viscosity as measured by the apparatus will be obtained from the relation

$$\eta = K.P.T,$$

where K = is the constant of the apparatus obtained by measurement with a liquid of known viscosity,
P = is the mean pressure acting on the liquid expressed in cms. of mercury column,
and T = is the time taken for out-flow expressed in seconds.

This is the relationship used in the present investigation.

The result obtained in the case of castor oil are given in Table VI, and curves of viscosity against T and $\log\eta$ against $1/T$ are shown in figures 8 and 9 respectively.

The densities and refractive indices of the oil under investigation were measured at the different temperatures and are given in Table I. The refractive indices were determined with an Abbe direct-reading refractometer provided with an arrangement for controlling the temperature. The densities were measured with a pyknometer and a sensitive chemical balance.

DISCUSSION

It is known that the absorption of gases by an oil takes place very slowly when it is kept at rest, and even when shaken the maximum absorption takes place after a period of time. In the present investigation, for each reading the shaking was continued till the maximum absorption was obtained, as indicated by the rise in mercury column in the manometer, the time taken being about 20 to 30 minutes, this period decreasing at higher temperatures.

The Bunsen absorption coefficient at different temperatures between the range 20°C—90°C were obtained by the method already described, and the results have been plotted in Figs. 2—5. The expansion of oil due to temperature rise has been taken into-consideration and the absolute values of absorption coefficient calculated. From the curves it will be seen that in the case of all the gases

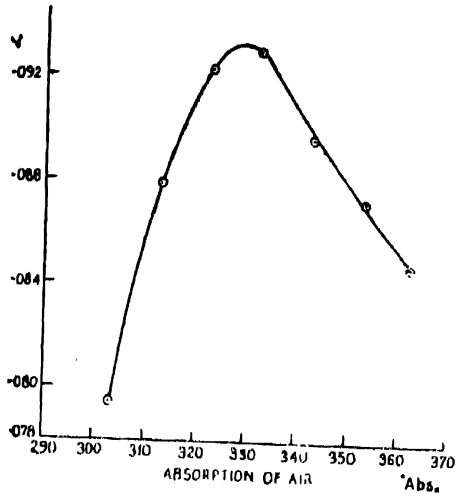

FIGURE 2

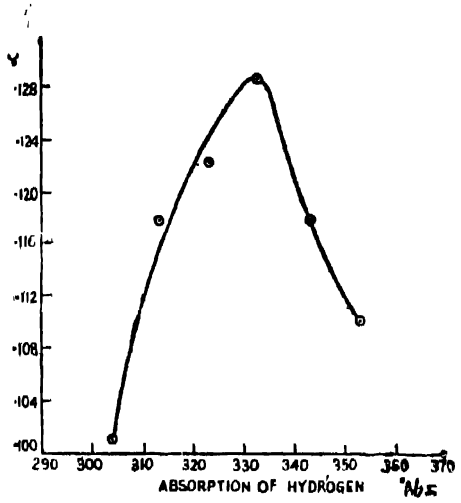

FIGURE 3

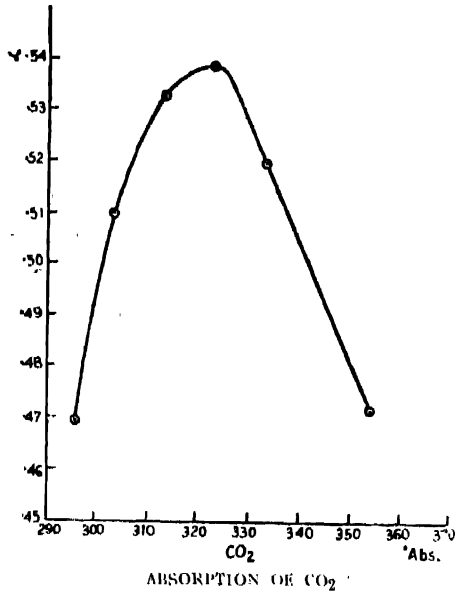

FIGURE 4

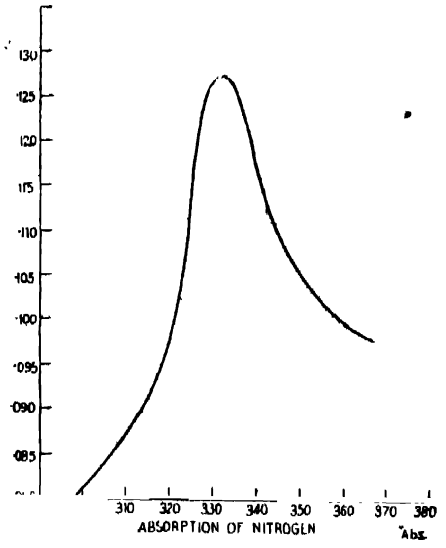

FIGURE 5

considered the absorption coefficient at first increases with temperature and after reaching a maximum value decreases with temperature. It will be seen that the nature of variation of absorption coefficient with temperature is almost similar for all the gases, the maximum absorption taking place at a temperature near about 60°C. The values obtained in the case of air and nitrogen are of the same order, perhaps because of the high percentage of nitrogen present in air. The values obtained in the case of CO_2 is much greater in comparison with those in the case of air, hydrogen and nitrogen. This is of particular interest on account of the fact that the structure of CO_2 (O=C=O) is almost similar to the chain-

like structure of the molecules of oil, and it is not known whether this higher absorption is due to a larger force of attraction between these two types of molecules.

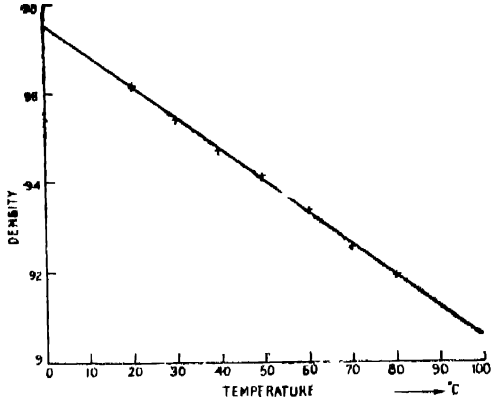

FIGURE 6

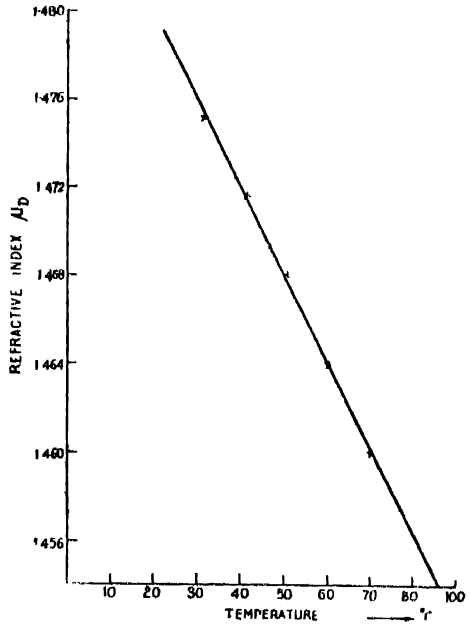

FIGURE 7

The variation of density and refractive index with temperature are shown in Figs. 6 and 7 respectively, and it will be seen that the variations are linear.

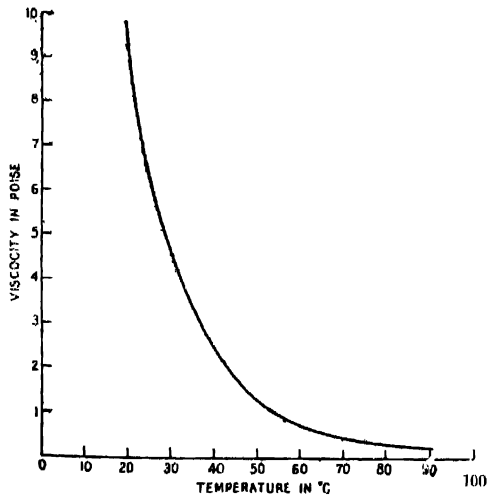

FIGURE 8

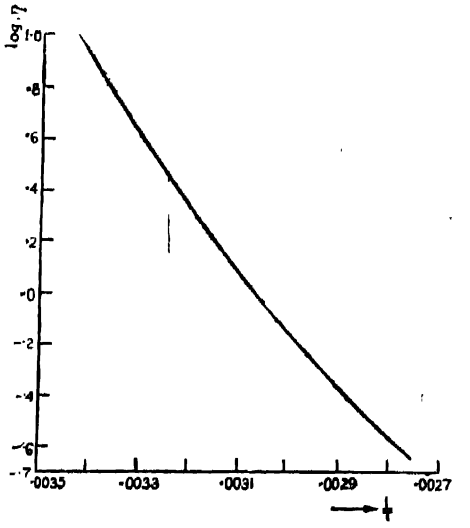

FIGURE 9

The variation of viscosity with temperature within the same range is shown in Fig. 8 and it will be seen that the viscosity decreases considerably with

increasing temperatures. The curve showing the change of logarithms of viscosity with $1/T$ is given in Fig. 9 and it will be observed that the curve is non-linear which shows that the variation of viscosity is due to some factor in addition to temperature. The logarithms of viscosity have been fitted by the method of least squares and the values of constants α, β, γ, δ, and log A have been worked out.

TABLE I

Data for Density and Refractive Index

(See Figs. 6, 7)

Temp. °C	Density	Refr. Index
20	.96122	1.4788
30	95396	1.4750
40	94704	1.4716
50	.94152	1.4680
60	.93380	1.4640
70	.92564	1.4600
80	.91973	--
90	91374	--

TABLE II

Data for Absorption of Air

Vol. per cm. of the tube = 0.16484 c.c. Vol. of oil = 49.45 c.c.

(See Fig. 2)

Temp. of oil in °C	Rise of mercury column in cm.	Vol. of gas absorbed in c.c.	Absorption Coeff. α	Density of oil	α corrected for vol. expansion of oil
20	—	—	—	.96122	—
30	27.0	4.451	.0811	.95396	.0794
40	31.1	5.128	.0904	.94704	.0879
50	33.9	5.589	.0955	.94152	.0923
60	35.5	5.862	.0970	.93380	.0930
70	35.5	5.862	.0942	.92564	.0895
80	35.8	5.902	.0923	.91973	.0871
90	35.8	5.902	.0987	.91937	.0847

TABLE III

Data for Absorption of Hydrogen

Vol. of oil = 49.4 c.c.

(See Fig. 3)

Temp. of oil in °C	Rise of mercury column in cm.	Vol. of gas absorbed in c c.	Absorption Coeff. α	Density of oil	α corrected for vol. expansion of oil
20	—	—	—	—	—
30	34 3	5.654	103	9533	.100
40	41·5	6.841	·120	·94704	.117
50	44·9	7.401	126	·94152	.122
60	49 0	8·078	.134	·93380	.128
70	46 8	7 714	·124	·92564	.118
80	45·3	7·467	116	·91973	·110
90	—	—	—	—	—

TABLE IV

Data for Absorption of Carbon-dioxide

Vol. of oil = 11.1 c.c.

(See Fig. 4)

Temp. of oil in °C	Rise of mercury column in cm.	Vol. of gas absorbed in c.c.	Absorption coeff. α	Density of oil	α corrected for Vol. expansion of oil
23	34·8	5·737	·476	·95904	·469
30	38.9	6.412	·520	95396	·509
40	42.3	6.970	·548	·94704	·532
50	44·4	7 319	·557	·94152	·538
60	44·5	7·336	540	·93380	519
70	45·0	7·401	·530	·92564	·504
80	43·5	7·171	·499	·919973	·471
90	—	—	—	—	—

TABLE V

Data for absorption of Nitrogen

Vol. of oil = 37.30 c.c.

(See Fig. 5)

Temp. of oil in °C.	Rise of mercury column in cm.	Vol. of gas absorbed in c.c.	Absorption coeff. α	Density of oil	α corrected for vol. expansion of oil
26.5	19.875	3.2751	0.084	0.9562	0.080
30	19.950	5.5382	0.087	0.9539	0.083
40	23.100	3.8067	0.003	0.9470	0.088
50	27.700	4.5650	0.108	0.9415	0.102
60	36.025	5.9345	0.137	0.9338	0.128
70	32.150	5.2987	0.118	0.9256	0.109
80	32.133	5.2956	0.114	0.9197	0.105
90	31.475	5.1375	0.108	0.9137	0.099

TABLE VI

Viscosity Data

(See Figs. 8, 9)

Temp. in °Abs.	η in poise.	$\log_{10}\eta$ (obs.)	$\frac{1}{T}\times 10^2$	$\log_{10}\eta$ (calc.)	obs. − cal.
293	0.82	.0921	·3413	.9904	·0017
303	4.53	.6561	·3301	.6827	.0034
313	2.31	.3636	·3192	·3600	.0036
323	1.28	.1072	·3096	.1073	·0001
333	0.769	−.1141	·3003	.1110	.0031
343	0.506	−.2958	.2915	·3006	.0048
353	0.344	−.4634	·2831	·4678	.0044
363	0.240	−.6198	·2755	.6190	.0008

$$\log_{10}\eta = \cdot 9904 - \cdot 3619\times 10^{-1}.\ t + 249\times 10^{-3}.\ t^2 - \cdot 776\times 10^{-6}.\ t^3 - 139\times 10^{-8}.\ t^4.$$

The measurements of absorption of different gases in the case of other oils are in progress and the results so far obtained agree with the observations made in this paper. Measurements on the conductivity of vegetable oils have also been taken up. These will be reported in a separate communication.

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